Pesticides in Ambient Air in Alberta

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FOREWORD

Pesticides in ambient air is a relatively unknown study area of environmental sampling, especially in Alberta. Extensive research has been done in Canada and elsewhere in the world on atmospheric concentrations of older organochlorine pesticides, which are known to be persistent and have been implicated in trans-boundary movements. Recent developments in analytical methodology and instrumentation have enabled the detection of lower levels of currently used pesticides in all environmental matrices. Along with the improvement in analytical technology, issues associated with air quality in general, and organic air contaminants in particular, have led to greater concerns about the quality of air in Alberta.

Alberta accounts for approximately 25% of the agricultural pesticides (herbicides, fungicides and insecticides) sold in Canada each year. As such, there is a need to characterize the impacts of this scale of usage on Alberta's environment. Several of these pesticides, such as triallate, trifluralin and ethalfluralin, are know to be volatile, to the extent that they have to be incorporated into soil immediately after application to minimize losses. Others, such as lindane and pentachlorophenol, are currently under review by regulatory agencies in Canada and the U.S. Still others, such as hexachlorobenzene, have not been used as pesticides for many years, but are still found as contaminants in some currently used pesticides, as well as being examined under international studies on persistent organic contaminants.

This project was undertaken under the direction of the Air Research Users Group of Alberta Environment to characterize the pesticides found in a number of Alberta locations and to determine their relative levels and seasonality. Information from this study will be used to assist in developing future research studies related to pesticide fate and pesticide presence in the atmosphere, assessing the need for ongoing monitoring of pesticides in Alberta's air, and providing valuable information on what organic contaminants such as currently used and historical pesticides can be found in Alberta's air. Data from this study, in conjunction with research currently being done by Agriculture and Agri-Food Canada in Lethbridge on pesticides in precipitation, will help provide Alberta Environment and other agencies with a more complete picture of the impact of pesticide use on Alberta's environment.

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SUMMARY

During this study pesticides were measured at four selected sites: Lethbridge, Vegreville, Lacombe and Lundbreck. These sites were chosen in consultation with Alberta Environment personnel based on various criteria including geographical location in the province, pesticide sales data from across the province and use patterns of various pesticides used in the province to control target insects, weeds and plant diseases. These sites also overlapped with Agriculture and AgriFood Canada pesticides in precipitation research sites. Lethbridge, Vegreville and Lacombe are agricultural areas while Lundbreck, the control site, is in the foothills of the Alberta Rockies. It was decided to monitor the residues through most of the year to cover as much of the weather regimen as possible.

Frequency of sampling on each site was also an important issue because of the time of application of pesticides in the various parts of the province as well as the variability in the weather conditions within the different crop growing areas. Most of the pesticides are applied from late April (seeding) to about early July to reflect the growing season and the target pest to be controlled. These kinds of pesticide application patterns are also necessary because some of the pesticides are applied pre-emergence while others are applied post emergence. Some selected pesticides are also applied quite late in the season just before the harvesting of the crop because of their use in pre-harvest weed control, although these pesticides were not analyzed in this study. Based on this kind of information the pesticide sampling frequency was scheduled so that more frequent samples were collected in June and July (almost weekly) while a biweekly schedule was chosen for the months of May, August and September. The frequency of sampling was slowed down to only once a month during October, November and January.

Interesting results were obtained when all the samples were analyzed using proper quality control and analytical procedures. Out of a target list of fifty-nine pesticides only nineteen pesticides were detected. Most pesticides were detected in the Lethbridge ambient air (19) followed by Vegreville (15), Lacombe (13) and Lundbreck (9). The pesticides that were detected at all the four sites include MCPA, bromoxynil, ethalfluralin, trifluralin, hexachlorobenzene (HCB), lindane, pentachlorophenol (PCP) and triallate. This closely compares to some of the data collected by Alberta Environment in a preliminary study during 1998 where trifluralin, triallate and ethalfluralin were almost always present at detectable levels. Compounds such as PCP and lindane have been found in ambient air in almost every part of the world where such measurements were carried out. In addition to the local use of these compounds they are also known to undergo long-range transport and have been detected in such remote places as the Arctic. Lindane and PCP are on list A-1 of some thirty compounds as decided by the Federal Reduction/Elimination of Toxics Program (ARET).

The Lethbridge sampling site did show some unique pesticides which were not detected at any other site and they include α-BHC and α-endosulfan. Some interesting observations can however be made from the results at this sampling site. Clopyralid which was only detected at Lethbridge and Vegreville was present at Lethbridge on June 15 while at the Vegreville site it was observed for about the first three weeks of July.

Interestingly MCPP, chlorpyrifos and pentachloronitrobenzene (PCNB) were also detected only at these two sites. This may actually be the result of application times of these compounds in these areas. PCNB was only found at both sites in June while MCPP was found around the end of June and the beginning of July only. Chlorpyrifos was detected at least once at Lethbridge during June, July and August while it was only detected once in July at Vegreville. The phenoxy herbicide 2,4-D was detected at Lethbridge, Lacombe and Vegreville only while dicamba was detected at Lacombe and Lethbridge. This is also quite consistent with some of the other monitoring studies carried out by Alberta Environment. Fenoxaprop was also detected at three sites, Vegreville, Lacombe and Lethbridge while malathion was detected at Vegreville, Lethbridge and Lundbreck.

Pesticides enter the atmosphere via drift, volatilization and wind erosion and are removed from the atmosphere via dry deposition and precipitation. More persistent compounds such as trifluralin, triallate, lindane, ethalfluralin, PCP and HCB can recycle numerous times in this manner. Some of the more volatile compounds such as trifluralin and triallate are not efficiently removed from the atmosphere via precipitation because they are not water soluble.
It should be pointed out that most probably due to cold climatic conditions and subzero temperatures during the month of January no pesticides were detected in samples collected from two of the sites. The only compounds detected were pentachlorophenol (0.03 ng/m^3) at the Vegreville site and hexachlorobenzene (0.02 ng/m^3) at the Lundbreck site.

PROJECT TEAM

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Gary Byrtus	AENV Project Coordinator

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1. INTRODUCTION

Total pesticide use in Alberta is estimated to be greater than 9300 tonnes of active ingredient annually. Once the pesticide has been used for its intended purpose, the fate of the pesticide in the environment becomes an important issue both from the provincial and national point of view. It is well known that there are several compartments of the environment that are impacted by pesticide use. There are three major environmental compartments, namely water, soil and air that are most important from the human health point of view. Pesticides have been monitored in surface waters and to a lesser extent, in ground water in Alberta for many years. The soil compartment has been sampled and monitored from time to time primarily for addressing the issues of soil contamination, accidental spills and poor crop performance. Little effort has been devoted to pesticide monitoring in air. With increasing focus on human exposure to pesticides from all sources including water, soil, food and air, interest in pesticide levels in ambient air in Alberta has increased. The ambient air pesticide levels data also provides a direct indication of widespread contamination of air resources with these air toxics. Assessment of impact of pesticides in the atmospheric environment is also complicated by the fact that virtually no guidelines exist for pesticides in ambient air. Most pesticide regulatory legislation deals with the impact of pesticide use on the aquatic environment and does not address the pesticide impact on the atmospheric environment adequately.

Pesticides enter the atmosphere via drift, volatilization and wind erosion and they are removed from the atmosphere via dry deposition and precipitation. More persistent pesticides can recycle numerous times in this manner and can transport over a large distance until they are finally broken down or trapped in glacial snowpacks. Although pesticides are classified as air toxics, they have not been addressed in any current integrated research projects and it was felt that necessary background data is required to enable Alberta Environment to initiate a review of this issue with a view to possibly include pesticides into the Alberta Air Toxics Program.

This research project was therefore approved by the Air Research Users Group in March 1999 with the primary objective of measuring and characterizing the number and concentration of pesticides found in Alberta air. The overall objectives of the project were to:

- Select four sampling sites across Alberta and conduct sampling on a weekly, biweekly and/or monthly basis for about a year based on the pesticide use at the selected sites.
- Draw a target list of pesticides to be monitored.
- Design a sampling protocol for all the sites.
- Set up the samplers at the sites and conduct the field sampling.
- Analyze the samples for pesticide target list.
- Evaluate the data and write a final client report.

2. MATERIALS AND METHODS

All reagents used throughout the study were pesticide grade. Standards were purchased as two certified mixes from Chromatographic Specialties. Glassware used throughout the study was thoroughly cleaned with soap and water followed by a rinse with deionized distilled water, acetone and dichloromethane. The glassware was then oven dried at 120°C.

The sampling sites were selected and chosen with the help of Alberta Environment personnel. After initial meetings, the four following sites were chosen for sampling.

1. Lethbridge (Ag. Canada research station) (49° 42' 19"; 112° 41' 44")
2. Vegreville (ARC site) (53° 30' 19"; 112° 05' 33")
3. Lacombe (farmstead 20 km SE of Lacombe) (52° 23' 17"; 113° 26' 43")
4. Lundbreck (1 km SW of Lundbreck Falls) (49° 33' 32"; 114° 13' 45")

The sites were selected based upon pesticide use intensity, site access and serviceability, and integration with a concurrent research study on pesticides in precipitation being carried out by Agriculture and Agri-Food Canada. Contacts were made with the appropriate personnel on all four sites and necessary permissions were obtained. Various field personnel who could help us in changing the filters and cartridges for each sample and carry out the sampling protocol were contacted. All the field personnel were trained in how to handle the equipment and were provided with written instructions to follow while changing and removing the PUF filter sampling heads. Necessary emergency contact names and phone numbers were also provided to facilitate and solve any complications that might arise during the sampling period. A sampling schedule for each site was decided upon and also given to the field personnel. An example of the sampling schedule for the Lethbridge site is given in Table 1. The sampling schedule was based upon anticipated spraying periods but was constrained by the funds allocated for analysis.

The method for sampling, analysis and determination of pesticides was adapted from the approved EPA method T010 which deals with determination of pesticides in ambient air. The air samplers used in this study were Graseby type PS-1 samplers which were calibrated using standard procedures at the Millwoods facility of Alberta Environment and the Vegreville facility of Alberta Research Council. Typical sampling times and sampling volumes were 24 hours and 300 - 400 m^3 of ambient air.

	Sunday	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday
					1	2	3
April	4	5	6	7	8	9	10
	11	12	13	14	15	16	17
	18	19	20	21	22	23	24
	25	26	27	28	29	30	1
May	2	3	4	5	6	7	8
	9	10	11	12	13	14	15
	16	17	18	19	20	21	22
	23	24	25	26	27	28	29
June	30	31	1	2	3	4	5
	6	7	8	9	10	11	12
	13	14	15	16	17	18	19
	20	21	22	23	24	25	26
	27	28	29	30	31	1	3
July	4	5	6	7	8	9	10
	11	12	13	14	15	16	17
	18	19	20	21	22	23	24
	25	26	27	28	29	30	31
Aug	1	2	3	4	5	6	7
	8	9	10	11	12	13	14
	15	16	17	18	19	20	21
	22	23	24	25	26	27	28
	29	30	31	1	2	3	4
Sept	5	6	7	8	9	10	11
	12	13	14	15	16	17	18
	19	20	21	22	23	24	25
	26	27	28	29	30	1	2
Oct	3	4	5	6	7	8	9
	10	11	12	13	14	15	16
	17	18	19	20	21	22	23
	24	25	26	27	28	29	30
Nov	31	1	2	3	4	5	6
	7	8	9	10	11	12	13
	14	15	16	17	18	19	20
	21	22	23	24	25	26	27
	28	29	30	1	2	3	4
Dec	5	6	7	8	9	10	11
	12	13	14	15	16	17	18
	19	20	21	22	23	24	25
	26	27	28	29	30	31	1
Jan	2	3	4	5	6	7	8
	9	10	11	12	13	14	15
	16	17	18	19	20	21	22
	23	24	25	26	27	28	29
	30	31	1	2	3	4	5
Feb	6	7	8	9	10	11	12
	13	14	15	16	17	18	19
	20	21	22	23	24	25	26
	27	28	29				

Table 1. Pesticide sampling dates at Lethbridge site 1999 - 2000

	Sunday	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday
					1	2	3
April	4	5	6	7	8	9	10
	11	12	13	14	15	16	17
	18	19	20	21	22	23	24
	25	26	27	28	29	30	1
May	2	3	4	5	6	7	8
	9	10	11	12	13	14	15
	16	17	18	19	20	21	22
	23	24	25	26	27	28	29
June	30	31	1	2	3	4	5
	6	7	8	9	10	11	12
	13	14	15	16	17	18	19
	20	21	22	23	24	25	26
	27	28	29	30	31	1	3
July	4	5	6	7	8	9	10
	11	12	13	14	15	16	17
	18	19	20	21	22	23	24
	25	26	27	28	29	30	31
Aug	1	2	3	4	5	6	7
	8	9	10	11	12	13	14
	15	16	17	18	19	20	21
	22	23	24	25	26	27	28
	29	30	31	1	2	3	4
Sept	5	6	7	8	9	10	11
	12	13	14	15	16	17	18
	19	20	21	22	23	24	25
	26	27	28	29	30	1	2
Oct	3	4	5	6	7	8	9
	10	11	12	13	14	15	16
	17	18	19	20	21	22	23
	24	25	26	27	28	29	30
Nov	31	1	2	3	4	5	6
	7	8	9	10	11	12	13
	14	15	16	17	18	19	20
	21	22	23	24	25	26	27
	28	29	30	1	2	3	4
Dec	5	6	7	8	9	10	11
	12	13	14	15	16	17	18
	19	20	21	22	23	24	25
	26	27	28	29	30	31	1
Jan	2	3	4	5	6	7	8
	9	10	11	12	13	14	15
	16	17	18	19	20	21	22
	23	24	25	26	27	28	29
	30	31	1	2	3	4	5
Feb	6	7	8	9	10	11	12
	13	14	15	16	17	18	19
	20	21	22	23	24	25	26
	27	28	29				

Table 1. Pesticide sampling dates at Lethbridge site 1999 - 2000

The Polyurethane Foam (PUF) cartridges used in the study were cut at ARC Vegreville using an in-house designed cutter. The PUF was purchased from a Calgary company and the cartridges were cut to fit the size of the sampler, which was a cylinder with a diameter of 2.25" and depth of 3.75". The PUF cartridges were cleaned by soxhlet extraction with acetone followed by a 16 hour extraction with 5% ether in hexane. After cleaning, the PUF cartridges were dried overnight under vacuum at room temperature and then loaded into the appropriate glass sampling cartridge that fit into the sampling head of the PS-1 sampler. The PUF cartridges were cleaned in batches of six. A representative PUF cartridge from each batch was proven clean by extracting it and analyzing the extract using the procedure described elsewhere in this report. The PUF cartridges were all stored under dark conditions as the exposure of cartridges to light changed their color slightly. The corresponding filters were cut to size in the laboratory from a batch of Pallflex filters purchased from Fluid Clarifications Inc. of Calgary, Alberta. After the samples were collected in the field, the sampling head was sealed and shipped intact to the laboratory where they were disassembled. The PUF cartridges and filters were stored together at -14°C in clean glass jars with screw cap lids, and analyzed when all PUF cartridges were received from a sampling event.

The PUF cartridges and the filters were extracted together using a soxhlet extractor. Using a pre-cleaned tong the cartridges and filters were removed from the sealed container and placed in a 300 mL Soxhlet extractor which has been previously rinsed with the extraction solvent, 5% ether in hexane. The round bottom flask of the extractor had 300 mL of the extraction solvent added to it. Precleaned glass boiling beads were also added to the flask. The samples were then extracted with approximately 4-6 cycles per hour for 16 hours. The solvent was then evaporated to approximately 0.5 mL using a Turbovap evaporator with the water bath at 35°C. 200 µL of hexane was then added to the tube followed by 1 mL of diazomethane solution. The contents of the tube were mixed well and the tube was allowed to stand in a fume hood for 10 minutes to complete the methylation. The methylated sample extract was then concentrated to less than 200 µL using a gentle stream of nitrogen. The extract was then quantitatively transferred to an injection vial with a 200 µL insert using a disposable pipette. The final volume was made up to 200 µL with hexane and 4 µL of a 20 ng/µL internal standard solution was added.

The extracts were analyzed using a Varian Ion Trap Mass Spectrometer. The appropriate gas chromatographic analytical conditions and the Varian Ion Trap operating parameters are listed below. Appropriate qualifier and quantitation ions were used for calculating concentrations of each pesticide analyte in the extract.

Capillary GC Analytical Parameters

Carrier Gas	Helium UHP: head pressure 12 psi
Column	30 m fused silica DB-5MSITD, 0.25 mm I.D., film thickness of 0.25 mm
Oven Temperature Program	Initial temperature 65°C, hold for 0.1 min and increase at 6°C/min to 300°C and hold for 2.24 min.
Injector	1078 temperature programmable split/splitless
Injection rate	5 µL/sec
Injector Temperature Program	Initial temperature 70°C, hold for 0.20 min and increase at 150°C/min to 300°C and hold for 41 min.
Injection volume	2 µL
Injector split/splitless program	0.00 min vent off 0.01 min vent on 1.50 min vent off

Mass Spectrometric Analytical Conditions (Varian Ion Trap)

Ion Trap Temperature	200°C
Transfer Line	275°C
Filament Current	20 μAmperes
Multiplier	standard auto tune voltage
Mass Range	70-399 m/z (for DFTPP use range 40-445)
Scan Rate	1.0 scan/sec
Scan Mode	Full scan
Run Time -	43 min
Scan Start Time	8.5 min
Background mass	60 m/z
Mass Defect	-5 mμ/100μ
Segments (m/z)	1: 10-99 tune factor 70 rf=37
	2: 100-190 tune factor 180 rf=37
	3: 191-399 tune factor 100 rf=37
	4: 400-650 tune factor 20 rf=37
AGC Target	65,000 counts

3. RESULTS

The sampling for pesticides in ambient air was conducted at four sites that are distributed across Alberta to represent various pesticide usages in southern and northern parts of Alberta. The four sites chosen were Vegreville, Lethbridge, Lacombe and Lundbreck. Lethbridge was selected as the southern site while Vegreville qualified as a northern site. Lacombe was selected as a central Alberta site and Lundbreck was the control site, being located in the Alberta foothills upwind of intensive agriculture. The samplers were installed at all four sites in the first week of April, 1999. The sampling schedule was decided upon and the sampling was conducted over a ten month period from April 1999 to January 2000. Over the whole sampling period seventeen samples were collected at each of the four sites. The samples were extracted and analyzed, as described elsewhere in this report, for a target list of 59 compounds. This target list of compounds and their corresponding method detection limits are given in Table 2. For the Lethbridge site, the sampling dates, sampling time and sample volumes are given in Table 3. The corresponding details for the sites for Lundbreck, Lacombe and Vegreville are given in Table 4, 5 and 6. The analytical results for pesticides in ambient air for the samples collected at the Lethbridge site are given in Table 7, and the results for the samples collected at Lundbreck, Lacombe and Vegreville sites are given in Tables 8, 9 and 10 respectively.

At the Lethbridge site, out of the 59 pesticides that were monitored, 19 pesticides were detected (Table 7) at least once during the sampling period. Two compounds, trifluralin and hexachlorobenzene, were detected in all the samples from April 11 to November 23, 1999. These compounds were not detected in the January 18, 2000 sample. The range of concentration for trifluralin was from a low of 0.01 ng/m^3 to a high of 0.14 ng/m^3 while the corresponding range of hexachlorobenzene was from 0.02 to 0.10 ng/m^3 with the exception of September 28, 1999 sample where a 0.67 ng/m^3 of hexachlorobenzene was detected. Ethalfluralin was also detected in all samples from April 11, 1999 to September 28, 1999 with a concentration range of 0.04 to 0.81 ng/m^3 (peak level detected in the June 22, 1999 sample). No ethalfluralin was detected after the end of September. Triallate was also detected in all the samples from April 11 to August 3, 1999. After the end of August, triallate was not detected until the October 26 sample at a low concentration of 0.07 ng/m^3. The peak concentration of triallate (1.07 ng/m^3) was detected on May 11, 1999 and the lowest concentration of triallate (0.07 ng/m^3) was observed on June 8, 1999. Pentachlorophenol was detected in 11 samples out of a total of 17 samples collected over the sampling period. No pentachlorophenol was detected in ambient air after the September 7, 1999 sample. A high value of 1.72 ng/m^3 was detected on August 3 and the lowest value of 0.04 ng/m^3 was present in ambient air on June 8, 1999. Bromoxynil was detected in all samples starting May 11 and peaking at 0.30 ng/m^3 level on June 15, 1999. From June 22 to August 3, bromoxynil was detected at a relatively steady concentration of 0.06 ng/m^3 level and slowly decreasing to not detected on August 17, 1999. Lindane was detected in ambient air at Lethbridge in all samples starting from May 11 to August 3. Lindane levels peaked on June 15 at 1.15 ng/m^3 while the low level of 0.23 ng/m^3 was present in ambient air on June 22, 1999. Substantial amounts of lindane were present in the air whenever it was detected in the sample. Other compounds were also detected in

ambient air at Lethbridge from time to time during the sampling season. Chlorpyrifos was detected in four samples with a peak level of 0.32 ng/m^3 on July 6, 1999. Detectable amounts 2,4-D were present in late May and early June and in the August 3 sample. Malathion was detected in two samples in July only with a relatively higher level of 0.78 ng/m^3 on July 6 and a level of 0.14 ng/m^3 on July 20, 1999.

Table 2. Target pesticides list and their Method Detection Limits (MDL)

Compound	MDL* (ng/m^3)	Compound	MDL (ng/m^3)
2,4,5-T	0.05	Ethalfluralin (Edge®)	0.05
2,4-D	0.05	Ethion	1.00
2,4-DB	0.05	Fenoxaprop-P-ethyl	0.40
2,4-DP	0.05	Guthion® (Azinphos – methyl)	2.00
Alachlor	0.05	Heptachlor	0.05
Aldrin	0.05	Heptachlor Epoxide	0.05
α-BHC	0.05	Hexachlorobenzene	0.05
α-Chlordane	0.05	Imazamethabenz-methyl (Assert®)	0.50
γ-Chlordane	0.05	Imazethapyr	0.20
α-Endosulfan	0.05	Imazamox	0.20
γ-BHC(Lindane)	0.05	Metolachlor	0.50
p,p'-Methoxychlor	0.03	Malathion	0.50
Atrazine	0.05	MCPA	0.05
β-Endosulfan	0.05	MCPB	0.20
Bromacil	0.30	MCPP (Mecoprop)	0.05
Bromoxynil	0.05	o,p-DDD	0.05
Carbathiin (Carboxin)	1.00	o,p-DDE	0.05
Chlorpyrifos (Dursban®)	0.05	o,p-DDT	0.05
Clopyralid (Lontrel®)	0.20	p,p'-DDD	0.05
Cyanazine	0.50	p,p'-DDE	0.05
Desethyl atrazine	0.50	Pentachloronitrobenzene	0.05
Desisopropyl atrazine	0.50	Pentachlorophenol	0.05
Diazinon	0.05	Phorate (Thimet®)	0.05
Dicamba (Banvel®)	0.20	Picloram (Tordon®)	0.05
Diclofop-methyl(Hoe-Grass®)	0.20	Pyridaben	0.20
Dieldrin	0.05	Quinclorac	0.05
Dimethoate (Cygon®)	0.05	Terbufos	0.30
Disulfoton (Di-Syston®)	2.00	Triallate (Avadex BW®)	0.05
Diuron	2.00	Trifluralin (Treflan®)	0.05
Endosulfan Sulfate	0.05		

*All reported MDL are based on a sample volume of 300 m^3. The detection levels are then adjusted to correspond to various total volumes of individual samples. Similarly, the reported values of pesticide detections are adjusted to reflect the different sample volumes.

Table 3. Pesticide ambient air samples collected at Lethbridge

Sampling Date	Sample Number	Sampling Time (Hrs)	Sample Volume (m^3)
April 11, 1999	T99-0662	23.98	359
May 11, 1999	T99-0979	23.96	346
May 25, 1999	T99-1157	23.79	368
June 01, 1999	T99-1198	24.00	365
June 08, 1999	T99-1330	--*	300*
June 15, 1999	T99-1350	16.94•	255
June 22, 1999	T99-1452	23.90	358
July 06, 1999	T99-1605	24.53	374
July 20, 1999	T99-1709	24.00	370
July 27, 1999	T99-1785	23.95	362
August 03, 1999	T99-1800	23.96	364
August 17, 1999	T99-1958	24.03	374
September 07, 1999	T99-2070	23.95	362
September 28, 1999	T99-2318	23.99	383
October 26, 1999	T99-2534	23.87	380
November 23, 1999	T99-2686	23.92	372
January 18, 2000	T00-0087	24.04	398

*The sampling pump malfunctioned. Sample volume estimated at 300 m^3.
• Sampling time was shorter as the pump was still malfunctioning.

Table 4. Pesticide ambient air samples collected at Lundbreck

Sampling Date	Sample Number	Sampling Time (Hrs)	Sample Volume (m^3)
April 22, 1999	T99-0773	24.53	409
May 13, 1999	T99-1051	24.44	403
May 27, 1999	T99-1163	24.49	393
June 03, 1999	T99-1230	24.43	403
June 10, 1999	T99-1331	24.52	417
June 17, 1999	T99-1402	24.43	408
June 24, 1999	T99-1469	24.47	407
July 8, 1999	T99-1613	24.46	404
July 22, 1999	T99-1711	24.40	402
July 29, 1999	T99-1787	24.45	408
August 05, 1999	T99-1875	24.40	402
August 19, 1999	T99-2025	24.41	402
September 09, 1999	T99-2146	24.57	424
September 30, 1999	T99-2382	24.49	412
October 28, 1999	T99-2539	24.49	411
November 25, 1999	T99-2687	24.50	412
January 20, 2000	T00-0103	24.49	420

Table 5. Pesticide ambient air samples collected at Lacombe

Sampling Date	Sample Number	Sampling Time (Hrs)	Sample Volume (m^3)
April 22, 1999	T99-0732	24.00	289
May 11, 1999	T99-0849	24.13	293
May 25, 1999	T99-1147	24.10	293
June 01, 1999	T99-1203	24.05	278
June 08, 1999	T99-1272	24.08	286
June 15, 1999	T99-1367	24.11	287
June 22, 1999	T99-1424	24.10	295
July 06, 1999	T99-1539	24.10	287
July 20, 1999	T99-1686	24.05	278
July 27, 1999	T99-1761	24.11	272
August 03, 1999	T99-1821	24.10	279
August 17, 1999	T99-1953	24.11	281
September 07, 1999	T99-2048	24.18	289
September 28, 1999	T99-2305	24.15	291
October 26, 1999	T99-2533	24.12	291
November 23, 1999	T99-2685	24.08	307
January 18, 2000	T00-0104	24.07	315

Table 6. Pesticide ambient air samples collected at Vegreville

Sampling Date	Sample Number	Sampling Time (Hrs)	Sample Volume (m^3)
April 18, 1999	T99-0707	24.00	326
May 11, 1999	T99-0844	24.18	353
May 25, 1999	T99-1092	24.07	335
June 01, 1999	T99-1162	24.08	323
June 08, 1999	T99-1229	23.97	346
June 15, 1999	T99-1332	23.90	345
June 23, 1999	T99-1425	24.07	352
July 06, 1999	T99-1504	24.05	347
July 20, 1999	T99-1681	23.98	374
July 27, 1999	T99-1710	24.06	339
August 03, 1999	T99-1786	24.03	347
August 17, 1999	T99-1902	23.92	341
September 07, 1999	T99-2039	24.04	349
September 28, 1999	T99-2243	24.00	349
October 26, 1999	T99-2535	24.04	355
November 23, 1999	T99-2684	23.84	360
January 18, 2000	T00-0076	23.81	368

Table 7. Pesticides in ambient air at Lethbridge (ng/m^3)

Compound	Sampling Dates																% Detections (N=17)	Range
	Apr 11	May 11	May 25	Jun 01	Jun 08	Jun 15	Jun 22	Jul 06	Jul 20	Jul 27	Aug 03	Aug 17	Sep 07	Sep 28	Oct 26	Nov 23	Jan 18		
MCPA			0.03	0.10														12	ND→0.10
Clopyralid						2.22												6	ND→2.22
Dicamba					0.05													6	ND→0.05
MCPP			0.04	0.15														12	ND→0.15
Bromoxynil		0.08	0.08	0.10	0.02	0.30	0.06	0.06	0.04	0.03	0.02							59	ND→0.10
2,4-D			0.05	0.04							0.10							18	ND→0.10
Ethalfluralin	0.66	0.56	0.04	0.69	0.16	0.53	0.81	0.19	0.23	0.56	0.72	0.11	0.03	0.12				82	ND→0.81
α-BHC				0.02														6	ND→0.02
Pentachloronitrobenzene					0.25		0.18											12	ND→0.25
Trifluralin	0.01	0.05	0.01	0.05	0.02	0.05	0.06	0.06	0.07	0.03	0.05	0.04	0.01	0.14	0.01	0.01		94	ND→0.14
Hexachlorobenzene	0.08	0.10	0.03	0.05	0.10	0.04	0.04	0.04	0.03	0.03	0.03	0.02	0.04	0.67	0.03	0.03		94	ND→0.67
Lindane		0.34	0.67	0.72	0.45	1.15	0.23	0.33	0.33	0.94	0.40							60	ND→1.15
Diazinon									0.27									6	ND→0.27
Pentachlorophenol	0.06		0.17	0.23	0.04	0.17		0.32	0.16	0.22	1.72	0.51	0.06					65	ND→1.72
Triallate	0.14	1.07	0.09	0.58	0.07	0.68	0.19	0.30	0.16	0.21	0.43				0.07			71	ND→1.07
α-Endosulfan										0.41								6	ND→0.41
Chlorpyrifos				0.02		0.10		0.32				0.06						24	ND→0.32
Malathion								0.78	0.14									12	ND→0.78
Fenoxaprop			0.03		0.02													12	ND→0.03

Blanks indicate compound not detected; ND-Not Detected

Table 8. Pesticides in ambient air at Lundbreck (ng/m^3)

Compound	Sampling Dates																% Detections (N=17)	Range
	Apr 22	May 13	May 27	Jun 03	Jun 10	Jun 17	Jun 24	Jul 8	Jul 22	Jul 29	Aug 05	Aug 19	Sep 09	Sep 30	Oct 28	Nov 25	Jan 20		
MCPA					0.06													6	ND→0.06
Bromoxynil		0.08	0.02	0.04														18	ND→0.08
Ethalfluralin	0.09																	6	ND→0.09
Trifluralin	0.09	0.01		0.01							0.01			0.10				35	ND→0.10
Hexachlorobenzene	0.11	0.06	0.02	0.04			0.04		0.03	0.02	0.02	0.02	0.19	0.49	0.03	0.01	0.02	82	ND→0.49
Lindane			0.03	0.19				0.08	0.11	0.10	0.16							35	ND→0.19
Pentachlorophenol	0.15	0.14	0.15	0.28		0.53	0.17	0.31	0.51	0.66	0.17	0.55	2.82					70	ND→2.82
Triallate	1.10			0.18			0.13	0.11						0.48	0.05			35	ND→1.10
Malathion	0.16																	6	ND→0.16

Blanks indicate compound not detected; ND-Not Detected

Table 9. Pesticides in ambient air at Lacombe (ng/m^3)

Compound	Sampling Dates																% Detections (N=17)	Range
	Apr 22	May 11	May 25	Jun 01	Jun 08	Jun 15	Jun 22	Jul 06	Jul 20	Jul 27	Aug 03	Aug 17	Sep 07	Sep 28	Oct 26	Nov 23	Jan 18		
MCPA	0.17					0.44												12	ND→0.44
Dicamba										0.06								6	ND→0.06
MCPP						0.37												6	ND→0.37
Bromoxynil			0.02	0.14		0.18			0.02									24	ND→0.18
2,4-D			0.01															6	ND→0.01
Ethalfluralin		0.10	0.02	0.04														18	ND→0.10
Trifluralin	0.12	0.26	0.17	0.28	0.12	0.07	0.23	0.21	0.22	0.41	0.31	0.56	0.04	0.36	0.04			88	ND→0.56
Hexachlorobenzene	0.06	0.08	0.03	0.06	0.06	0.06	0.06	0.05	0.05	0.04	0.03	0.05	0.07	0.35	0.02			88	ND→0.35
Lindane		2.84	1.30	1.16	0.64	0.80	0.28	0.64	0.50	0.45	0.43	0.34						65	ND→2.84
Diazinon						0.12												6	ND→0.12
Pentachlorophenol	0.42	0.15	0.26	0.16	0.07	0.41	0.12	0.55	0.61	0.90	0.59	0.14						71	ND→0.90
Triallate	0.64	1.07	0.83	1.07	0.62	0.25	0.30	0.54	0.53	0.61	0.54	0.64			0.06			76	ND→1.07
Fenoxaprop					0.01	0.06												12	ND→0.06

Blanks indicate compound not detected; ND-Not Detected

Table 10. Pesticides in ambient air at Vegreville (ng/m^3)

Compound	Sampling Dates																% Detections (N=17)	Range
	Apr 18	May 11	May 25	Jun 01	Jun 08	Jun 15	Jun 23	Jul 06	Jul 20	Jul 27	Aug 03	Aug 17	Sep 07	Sep 28	Oct 26	Nov 23	Jan 18		
MCPA						0.46		0.19										12	ND→0.46
Clopyralid								0.34	0.13	0.07								18	ND→0.34
MCPP						0.27		0.28										12	ND→0.28
Bromoxynil						0.20		0.02										12	ND→0.20
2,4-D			0.01			0.36												12	ND→0.36
Ethalfluralin	0.03	0.04	0.09	0.04	0.22		0.02	0.08	0.11	0.12	0.06	0.16	0.02		0.01			76	ND→0.22
Pentachloronitrobenzene							0.28											6	ND→0.28
Trifluralin	0.12	0.40	0.31	0.22	0.39	0.02	0.04	0.13	0.32	0.30	0.10	0.26	0.07	0.05	0.02			88	ND→0.40
Hexachlorobenzene	0.07	0.13	0.04	0.05	0.06		0.06	0.05	0.03	0.03	0.03	0.03	0.04	0.44	0.02	0.01		88	ND→0.44
Lindane		0.54	2.92	1.51	1.17	1.38	0.28	0.68	0.87	0.75	0.43	0.49	0.09					71	ND→2.92
Pentachlorophenol	0.06	0.15	0.17	0.15	0.21	0.96	0.09	0.20	0.27	0.49	0.29	0.29	0.04				0.03	82	ND→0.96
Triallate	0.74	0.68	0.44	0.62	0.43	0.08	0.08	0.18	0.29	0.32	0.11	0.24	0.05		0.03			82	ND→0.74
Chlorpyrifos													0.02					6	ND→0.02
Malathion									0.13									6	ND→0.13
Fenoxaprop						0.17												6	ND→0.17

Blanks indicate compound not detected; ND-Not Detected

Pentachloronitrobenzene was detected in two samples only on June 8 (0.25 ng/m^3) and June 22 (0.18 ng/m^3). MCPA was detected in May 25 and June 1 samples only at a level of 0.03 and 0.10 ng/m^3 level. Fenoxaprop was detected on May 25 at 0.03 ng/m^3 and on June 8 at 0.02 ng/m^3 level only. Clopyralid was detected at a relatively higher level of 2.22 ng/m^3 on June 15 and was not detected in any of the other samples throughout the sampling period. Other compounds that were detected only in one sample during the sampling period include α-endosulfan (at 0.41 ng/m^3 level on July 27), diazinon (at 0.27 ng/m^3 on July 20), α-BHC (at 0.02 ng/m^3 level on June 1) and dicamba (at 0.05 ng m^3 level on June 01, 1999).

A problem with the motor of the sampler at Lethbridge on the June 8 sampling date likely resulted in lower than actual concentrations being reported for this sampling date.

At the Lundbreck site only nine pesticides were detected (Table 8) at least once during the sampling period. Hexachlorobenzene was the most frequently detected compound at this site and was detected in all samples except a couple of samples during the middle of June. The relative amount of hexachlorobenzene in ambient air was small, in the range of 0.01 to 0.11 ng/m^3, with the exception of a relatively high level of 0.49 ng/m^3 seen on September 30. Pentachlorophenol was detected in all samples from April 22 to September 9 with a relatively high value of 2.82 ng/m^3 on September 9 while at all the other times the concentration levels were in the range of 0.14 to 0.66 ng/m^3. Trifluralin was also detected in five samples on April 22, May 13, June 3, August 5 and September 30. Only on April 22 and September 30 were the trifluralin levels elevated at approximately 0.10 ng/m^3 while at the other three times the levels were rather low at 0.01 ng/m^3. Triallate was detected in six samples, on April 22, June 3, June 24, September 30 and October 28. The higher levels of triallate at 1.10 ng/m^3 and 0.48 ng/m^3 were detected on April 22 and September 30, respectively, while a lower level of 0.05 ng/m^3 was present on October 28. Lindane was detected in six samples in late May, early June, late July and early August. Lindane concentrations were higher on June 3 (0.19 ng/m^3) and August 5 (0.16 ng/m^3) than on May 27 (0.03 ng/m^3) and July 29 (0.01 ng/m^3). MCPA was detected only once during the sampling period on June 10 at a concentration level of 0.06 ng/m^3. Bromoxynil was observed in the ambient air at this site from the middle of May to early June and the range of concentration in this case was from 0.02 to 0.08 ng/m^3. Ethalfluralin was detected on April 22 at a 0.09 ng/m^3 level but was not present in any samples after that. Malathion was also observed in the April 22 sample at a concentration level of 0.16 ng/m^3 but was not present in any of the subsequent samples.

At the Lacombe site 13 pesticides were detected (Table 9) at least once during the sampling period. Trifluralin and hexachlorobenzene were detected in all samples from April 22 to October 26. Trifluralin levels in the ambient air peaked at 0.56 ng/m^3 on August 17 while the lowest level of 0.04 ng/m^3 was detected on September 7 and October 26. Hexachlorobenzene levels remained relatively steady at 0.04 to 0.06 ng/m^3 from April 22 to October 26 with the exception of the September 28 sample where a concentration level of 0.35 ng/m^3 was detected for hexachlorobenzene. Triallate was detected in ambient air in all samples from April 22 through to August 17 with the peak

concentration of 1.07 ng/m^3 observed on May 11 and June 1 while a low level of 0.06 ng/m^3 was observed on October 26.

Elevated levels of triallate (0.25 to 1.07 ng/m^3) were present in ambient air at this site from April to the middle of August. Pentachlorophenol was observed in all samples from April 22 to August 17 with a peak value of 0.90 ng/m^3 detected on July 27 and a low value of 0.07 ng/m^3 observed on June 8, 1999. Pentachlorophenol at this site also appeared to follow a similar trend as detected at the Lethbridge and Vegreville sites. Lindane was detected in all the samples from May 11 to August 17 with a peak value of 2.84 ng/m^3 on May 11 and a low value of 0.28 ng/m^3 on June 22. Elevated levels of lindane were present in ambient air from the beginning of May to the middle of August sampling period. Ethalfluralin was detected at this site only in three samples from May 11 to June 1 with a concentration range of 0.02 to 0.10 ng/m^3. Bromoxynil was present during the May 25 to June 15 sampling season with a higher level of 0.18 ng/m^3 on June 15 and a lower level of 0.02 ng/m^3 on May 25. MCPA was detected in two samples only, on April 22 at a level of 0.17 ng/m^3 and on June 15 at a 0.44 ng/m^3 level. MCPP was detected only in one sample on June 15 at a 0.37 ng/m^3 level. Dicamba and 2,4-D were each detected only once with a level of 0.06 ng/m^3 on July 27 for dicamba and a level of 0.01 ng/m^3 on May 25 for 2,4-D. Fenoxaprop was detected in June only twice at the concentration range of 0.01 to 0.06 ng/m^3. Diazinon was detected only once at 0.12 ng/m^3 on June 15.

At the Vegreville site, fifteen pesticides were detected (Table 10) at least once during the sampling period. No pesticides were detected at this site on the January 18, 2000 sampling date except for pentachlorophenol (0.03 ng/m^3). Trifluralin and hexachlorobenzene were detected in all samples from April 18 to October 26. Only hexachlorobenzene was detected in the November 23, 1999 sample. Trifluralin peaked at a level of 0.40 ng/m^3 on May 11 while a low value of 0.02 ng/m^3 was detected on June 15 and October 26, 1999. Hexachlorobenzene concentrations remained relatively steady (0.03 to 0.05 ng/m^3) during most of the sampling season with the exceptionally high value of 0.44 ng/m^3 on September 28, 1999. Pentachlorophenol and triallate were detected in all the samples from April 18 to September 7. The peak value of 0.96 ng/m^3 of pentachlorophenol was detected on June 15 while the lowest value of 0.04 ng/m^3 was detected on September 7, 1999. Triallate followed a similar pattern at this site with a peak value of 0.74 ng/m^3 on April 18 and a low value of 0.03 ng/m^3 on October 26, 1999. Relatively higher values of triallate were present on April 18, through to June 8 and July 20 to August 17 while relatively lower values were detected on June 15 and 23 and September 7, 1999. Lindane was detected in ambient air at this site in all samples from May 1 to September 7, 1999 with a peak value of 2.92 ng/m^3 detected on May 25, 1999 and a low value of 0.09 ng/m^3 detected on September 7, 1999. Elevated levels of lindane were detected at this site during the months of May, June, July and August. Ethalfluralin was detected in most of the samples from April 18 to October 26, although the relative range of concentration variation was only from 0.01 ng/m^3 to 0.22 ng/m^3. Clopyralid was detected in three samples during the month of July with a peak value of 0.34 ng/m^3 on July 6 and declining to 0.07 ng/m^3 by July 27, 1999. MCPA and MCPP were also detected in two samples each, once on June 15 and then on July 6, 1999. MCPA was

present at 0.46 ng/m^3 on June 15 and at a 0.19 ng/m^3 on July 6, 1999. MCPP was detected at a level of 0.28 ng/m^3, both on June 15 and July 6, 1999. Bromoxynil was detected at a level of 0.20 ng/m^3 on June 15 and 0.02 ng/m^3 on July 6. 2,4-D was detected at this site on June 15 at a relatively high level of 0.36 ng/m^3 while a low concentration of 0.01 ng/m^3 was present on May 25, 1999. Pentachloronitrobenzene was detected only in one sample on June 23, 1999 at a 0.28 ng/m^3 concentration level. Chlorpyrifos was present in ambient air on September 7 at a concentration of 0.02 ng/m^3. Malathion and fenoxaprop were each detected only once, on July 20 (0.13 ng/m^3) and on June 15 (0.17 ng/m^3) respectively.

4. DISCUSSION

Some of the interesting issues that arise from the results obtained under this project deal with the persistence of the pesticides lindane, hexachlorobenzene (HCB) and pentachlorophenol (PCP).

Lindane levels are high in the month of May and then slowly decline through the summer months. No lindane was detected in the samples in April and the levels had gone down to not detectable by the end of September (Figure 1). As lindane is used on treated seed that is planted in April and early May, lindane is then released into the atmosphere following seeding and hence the higher levels in May followed by a slow decline to low and/or undetectable levels in August and September.

Hexachlorobenzene levels seem to be rather steady in the range of ND 0.1 to ng/m^3 from April to August. However, the HCB levels rise to 0.6 ng/m^3 in September and again decline to < 0.1 ng/m^3 by October. It is not immediately apparent as to why this spike in HCB levels in September (Figure 2) is detected. However, a couple of possible explanations can be advanced as follows. HCB is a known contaminant of some fungicides currently in use (e.g. pentachloronitrobenzene - PCNB) and may have been released from the use of PCNB. It must however be noted that PCNB was itself detected in only three samples in the month of June. Another possibility is that HCB is not a local contaminant but something that arrived in Alberta as a result of long-range transport of HCB from elsewhere. This argument is partly supported by data provided by Environment Canada where the 5-day back trajectory plots showed the origins of the air mass sampled in Alberta in late September to be from the Bering Sea. Both of these hypotheses are not fully supported by this current study and more research work in some detail will be required to address this issue. Another interesting compound that somewhat resembles HCB in behaviour is pentachlorophenol (PCP). PCP is also a rather persistent compound that is detected at a level of ND to 0.5 ng/m^3 in April and May. PCP levels slowly rose in the month of June and July to 0.9 ng/m^3 levels and peaked in August and September at a level of 2.7 ng/m^3. The levels of PCP in ambient air quickly decreased to trace and/or ND by October and November (Figure 3). Once again based on the current study it is hard to suggest any plausible explanation for this spike of PCP in August and September. Once again it may be present in the air at this time because of long-range transport of PCP from elsewhere.

Some of the pesticides monitored during this study follow the expected trend that matches their time of application and subsequent release into the atmosphere. Triallate, for example, is applied as a pre-emergence herbicide and therefore one would expect the highest levels of triallate in ambient air in April and May. This matches the observed results to a large extent.

Based on the frequency of detections (depicted as percent detections based on a data set of 17 determinations in Tables 7-10) it is clear that the pesticides of interest in the ambient air include HCB, lindane, PCP, triallate and ethalfluralin as they are detected most frequently. The pesticides on the lower frequency of detection scale include MCPA, dicamba, malathion and endosulfan which are detected in only a couple of samples. Another possible explanation that has been used from time to time in the literature is "cycling" of some of these pesticides in the environment. The pesticides, once applied to the crop for their intended use can recycle itself between the air and other phases (soil and water) during the season. This will result in the air levels of pesticides to sort of start at a detectable level and then go up and down during the summer and slowly decline to trace or non-detectable levels by October. During this study some of the compounds that might fit this scenario include ethalfluralin, lindane, trifluralin, PCP and triallate. Other factors that will have an impact on the presence of these compounds in ambient air are vapour pressure and long-range transport of these pesticides. In order to address this issue with some confidence we need to study the residues in soil and water simultaneously with air, and possibly relate future work with long-range transport studies.

Some of the other variables that will have an impact on this issue include effect of temperature and effect of moisture as it relates to the release of these pesticides into the ambient air. Some of the questions that still require further investigation include why did the PCP and HCB concentrations spike in September; do we need long term air monitoring of pesticides; and can we use air sampling to measure the effect of use reductions of certain pesticides (such as lindane from canola seed treatment) in estimating environmental loading?

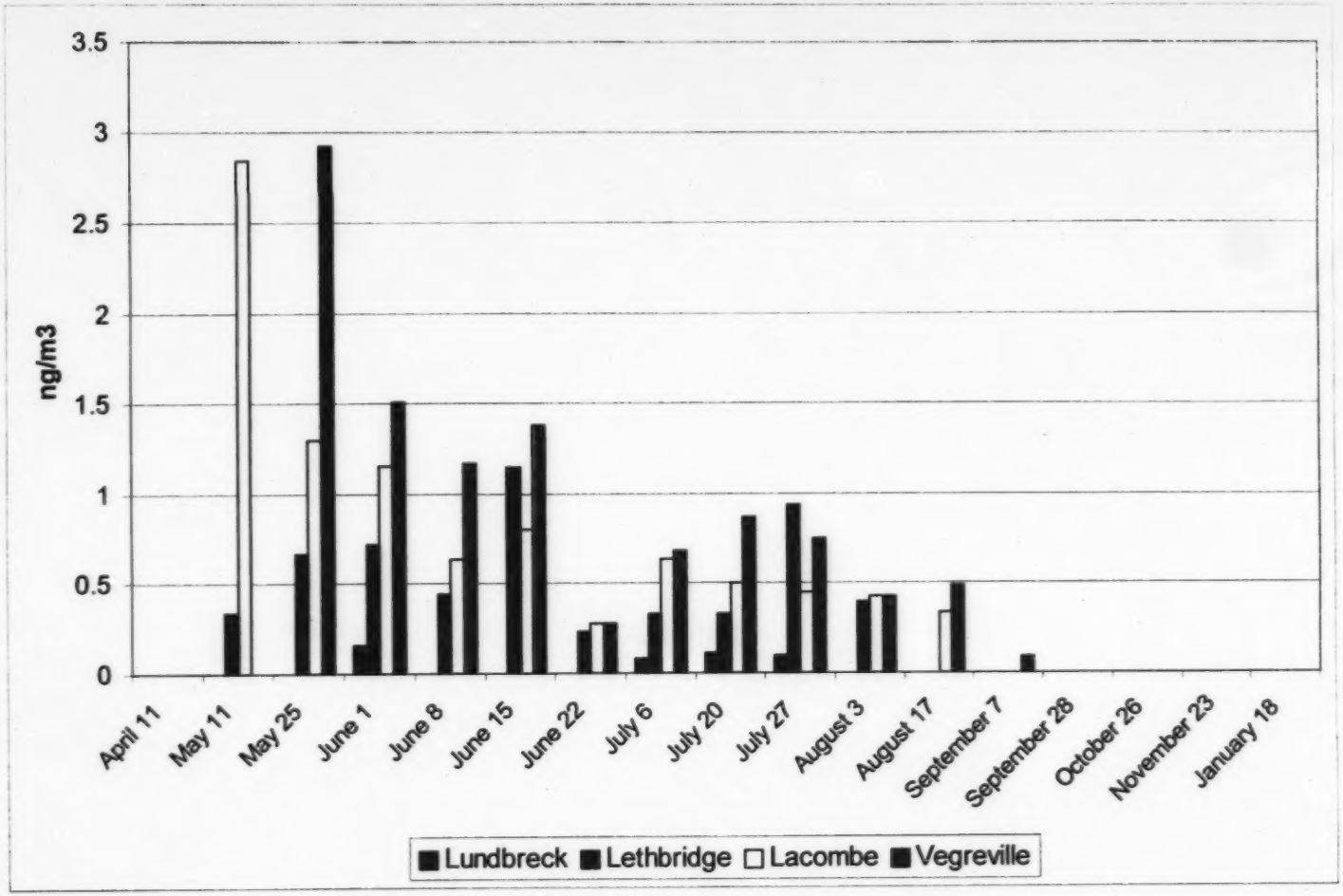

Figure 1. Concentration of Lindane in Alberta ambient air – 1999 – 2000

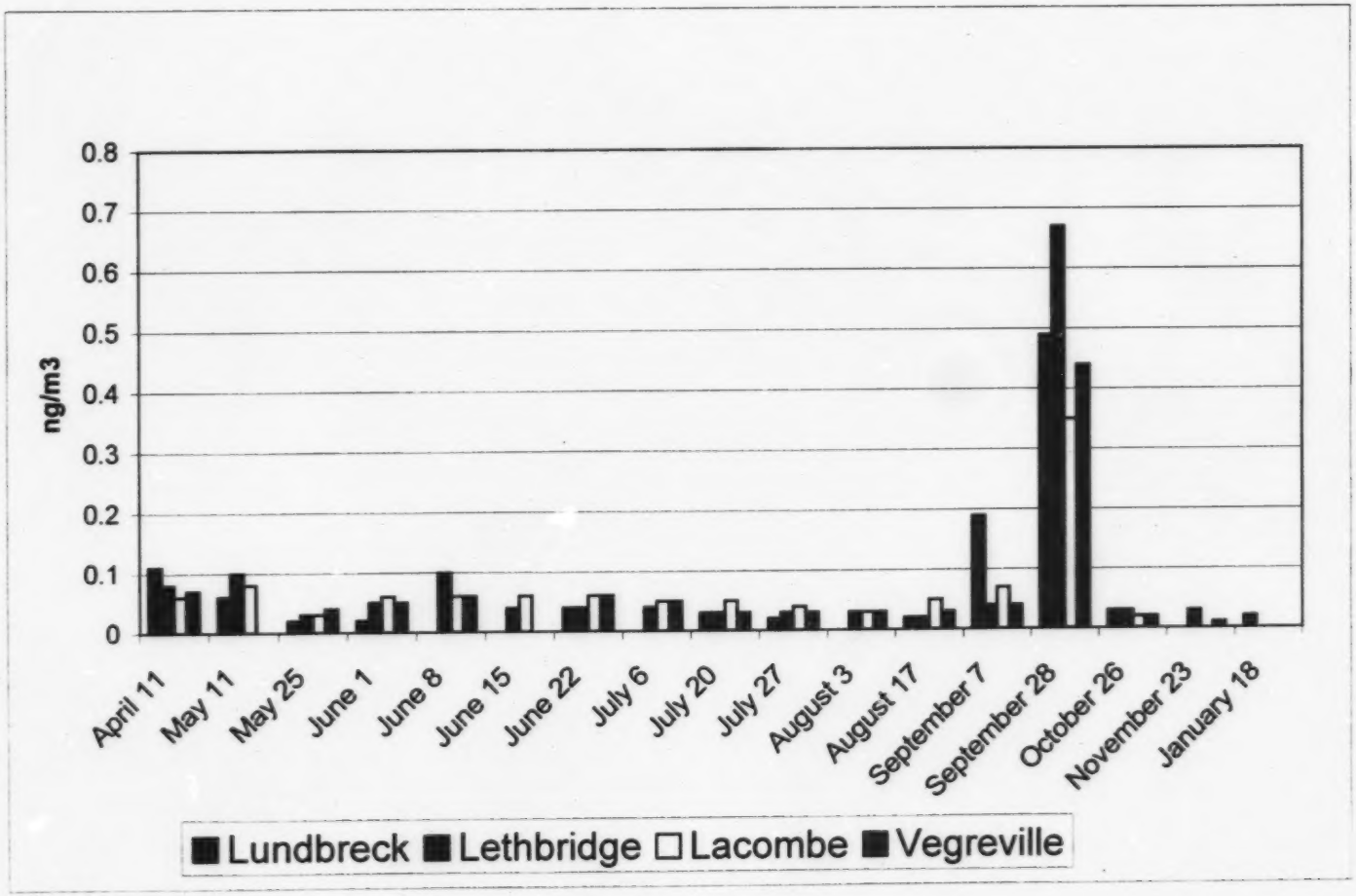

Figure 2. Hexachlorobenzene concentrations in air – 1999 – 2000

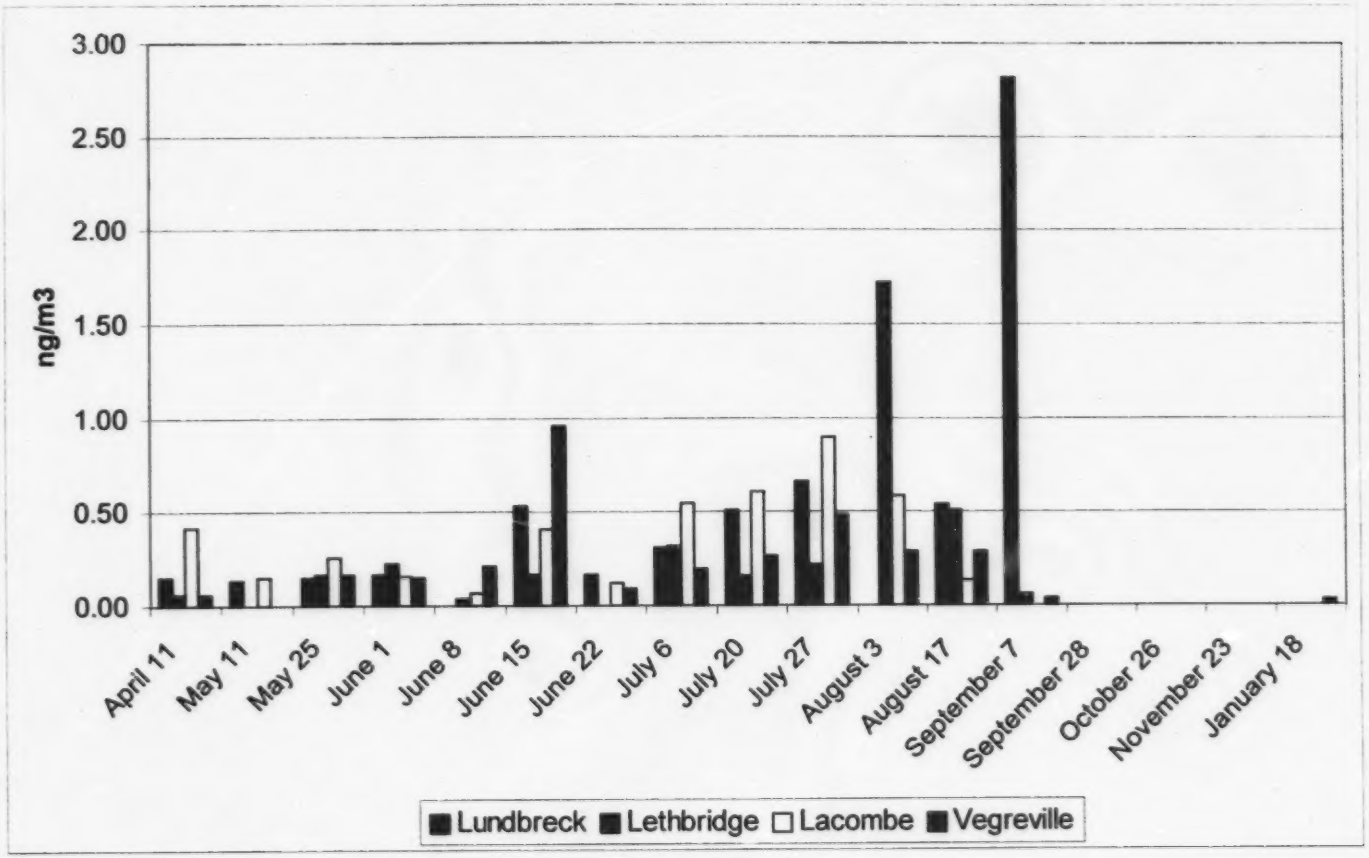

Figure 3. PCP concentrations in ambient air – 1999-2000

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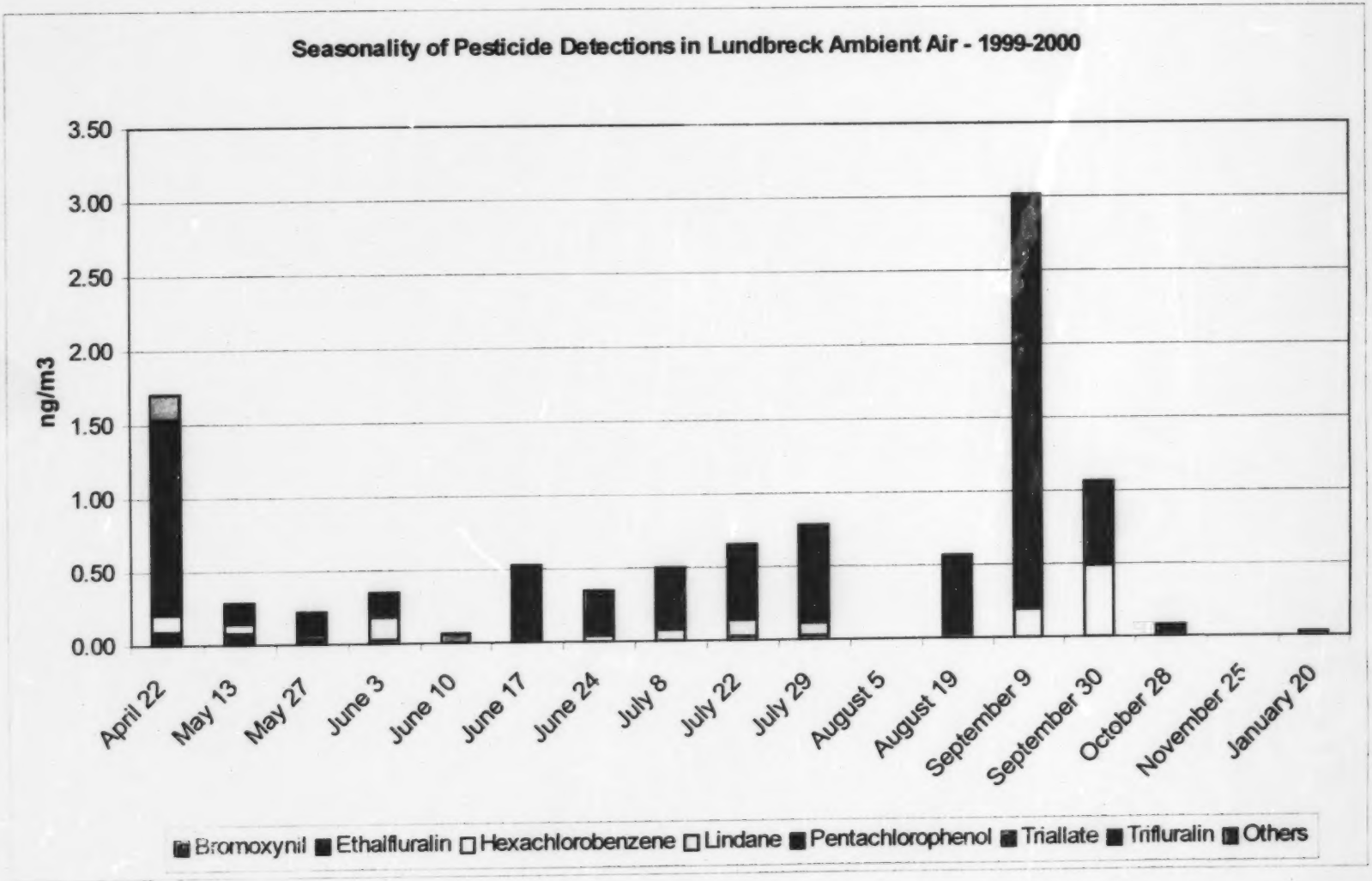
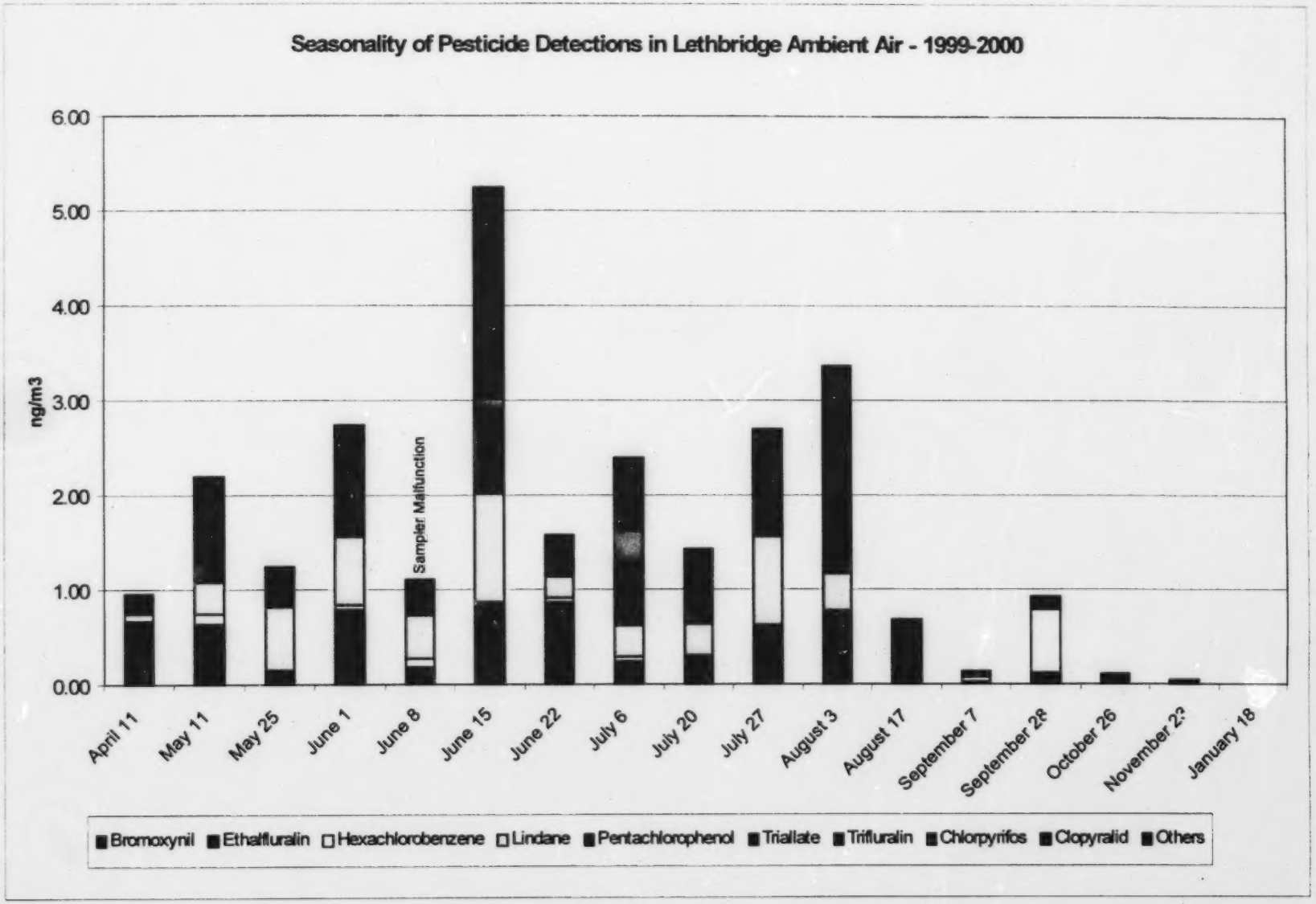
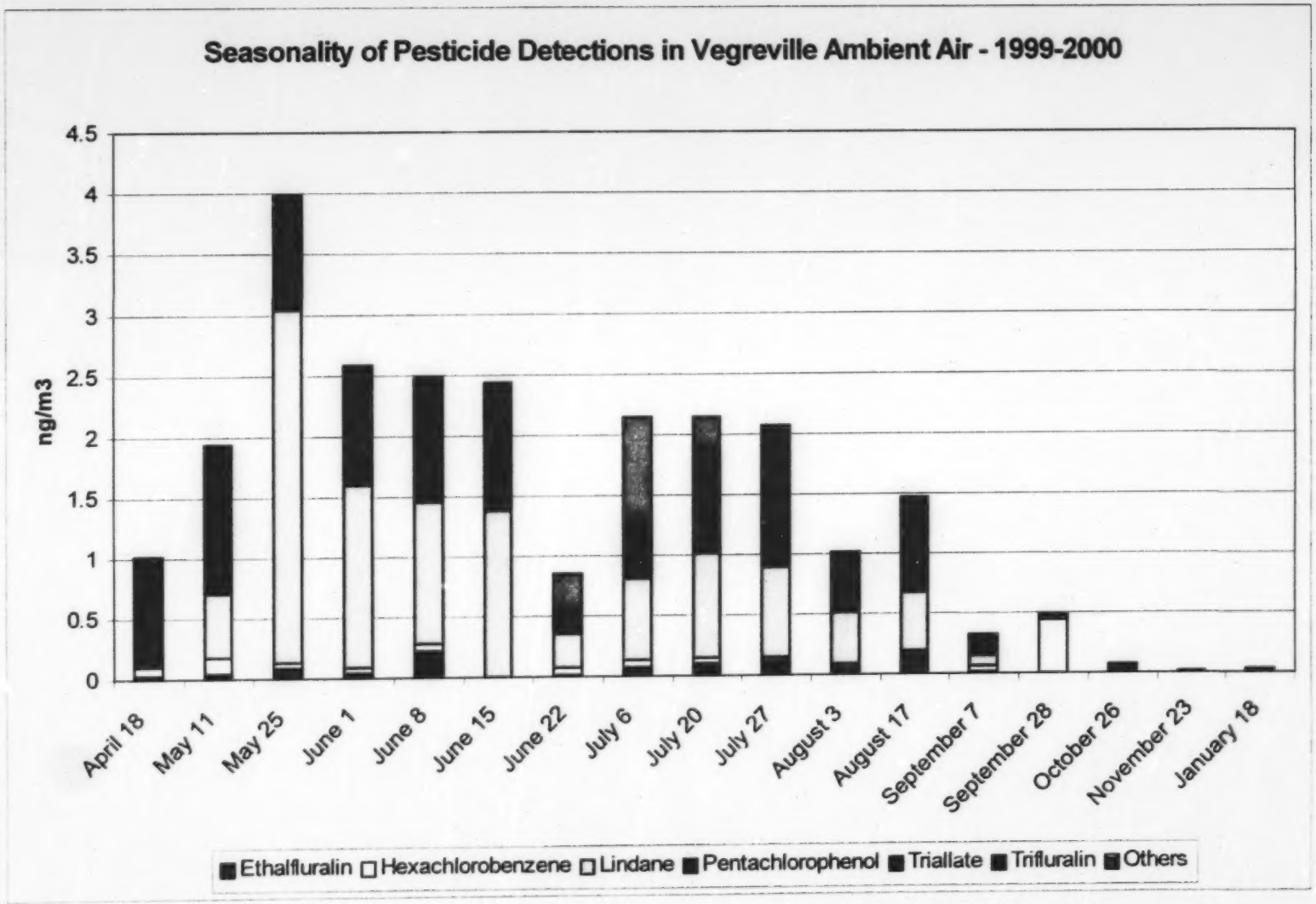
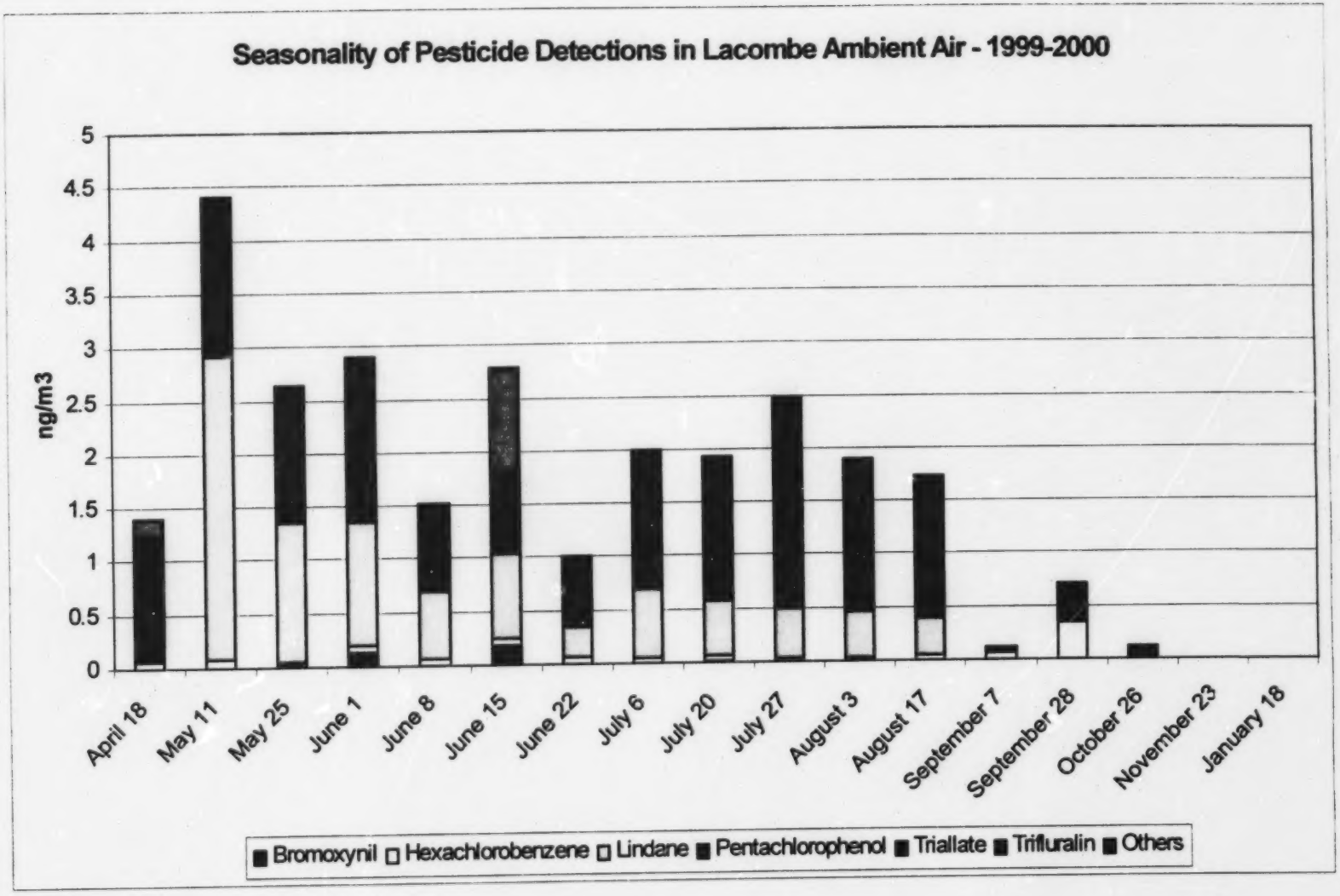
6. APPENDICES

Graphical Representation of Pesticides in Ambient Air Data


Seasonality of Pesticide Detections in Lundbreck Ambient Air - 1999-2000
ng/m3
3.50
3.00
2.50
2.00
1.50
1.00
0.50
0.00
April 22
May 13
May 27
June 3
June 10
June 17
June 24
July 8
July 22
July 29
August 5
August 19
September 9
September 30
October 28
November 25
January 20
Bromoxynil
Ethalfluralin
Hexachlorobenzene
Lindane
Pentachlorophenol
Triallate
Trifluralin
Others


Seasonality of Pesticide Detections in Lethbridge Ambient Air - 1999-2000
ng/m3
6.00
5.00
4.00
3.00
2.00
1.00
0.00
Sampler Malfunction
April 11
May 11
May 25
June 1
June 8
June 15
June 22
July 6
July 20
July 27
August 3
August 17
September 7
October 26
January 18
Bromoxynil
Ethalfluralin
Hexachlorobenzene
Lindane
Pentachlorophenol
Triallate
Trifluralin
Chlorpyrifos
Clopyralid
Others


Seasonality of Pesticide Detections in Vegreville Ambient Air - 1999-2000
ng/m3
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
April 18
May 11
May 25
June 1
June 8
June 15
June 22
July 6
July 20
July 27
August 3
August 17
September 7
September 28
October 26
November 23
January 18
Ethalfluralin
Hexachlorobenzene
Lindane
Pentachlorophenol
Triallate
Trifluralin
Others


Seasonality of Pesticide Detections in Lacombe Ambient Air - 1999-2000
ng/m3
5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
April 18
May 11
May 25
June 1
June 8
June 15
June 22
July 6
July 20
July 27
August 3
August 17
September 7
September 28
October 26
November 23
January 18
Bromoxynil
Hexachlorobenzene
Lindane
Pentachlorophenol
Triallate
Trifluralin
Others